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MISCIBILITY OF HYDROGEN AND HELIUM MIXTURES AT MEGABAR PRESSURES

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Models of Jupiter and Saturn postulate a central rock core surrounded by a fluid mixture of hydrogen and helium. These models suggest that the mixture is undergoing phase separation in Saturn but not Jupiter. State-of-the-art total energy calculations of the enthalpy of mixing for ordered alloys of hydrogen and helium confirm that at least partial phase separation has occurred in Saturn and predict that this process has also begun in Jupiter.

Keywords: Giant planets, hydrogen/helium miscibility

Evolutionary models determine the energy output of the planet as a function of time. These models yield a cooling rate for Jupiter which is consistent with both its measured energy output and the approximate age of the solar system, but the corresponding rate for Saturn is too fast.[1] It has been suggested[2] that as the planets cooled, a temperature might have been reached at some distance from the central core where helium-rich droplets would condense out of the hydrogen-helium mixture (phase separate), releasing gravitational energy as they settled. This extra source of energy might then account for the anomalously slow cooling of Saturn. However, it has been assumed that this process has not occurred in Jupiter.[1] To date the only internal energy source included in evolutionary models is the heat generated as a result of the gravitational collapse which occurred during the formation of the planet.[1] Calculations including the possibility of phase separation have yet to be carried out.

At the pressures in the interiors of Jupiter and Saturn, hydrogen is believed to be a liquid metal[3] while helium should be a wide band-gap insulator.[4] In spite of this fact, most previous calculations for hydrogen/helium mixtures start with a fully pressure-ionized system of ions and include the response of the electron gas using perturbation theory.[5-8] Using this approach, Stevenson[5] predicted that a 7% helium mixture would phase separate at approximately 7000 K near 8 Mbar. However, MacFarlane[9] predicted that hydrogen-helium mixtures would not phase separate at any temperature for

pressures near 10 Mbar. He used a model potential extracted from Thomas-Fermi-Dirac calculations for alloys of hydrogen and helium in bcc and fcc crystal structures.

State-of-the-art total energy (TE) methods used in condensed matter theory offer the possibility of an accurate description of both constituents. The accuracy of these methods, which are based on the local density approximation (LDA),[10] is well established.[11]

The primary result of our TE calculations (a more detailed account of the method and results will be published separately[12]) is given in Fig. 1 which shows the enthalpy of mixing per atom at 0 K and 10.5 Mbar,

$$\Delta H(x) = H(x) - xH(x=1) - (1-x)H(x=0), \tag{1}$$

where $H(x) = E(x) + PV(x)$ is the enthalpy per atom and x is the number fraction of helium atoms [E(x) is the total energy per atom, P is the pressure, which is held constant, and V(x) is the volume per atom]. In Fig. 1, the label sc refers to the calculations carried out for a simple cubic unit cell in both bcc (2 atoms/cell) and fcc (4 atoms/cell) lattices. Similarly, dia refers to a

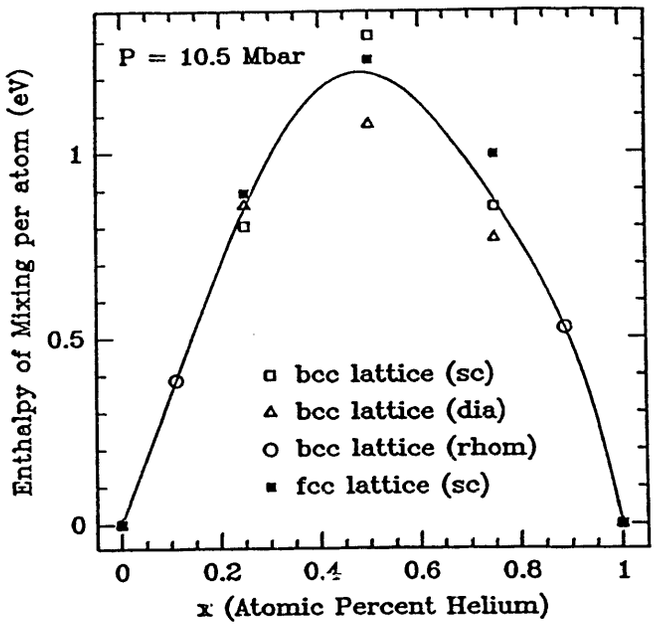

Fig. 1. The enthalpy of mixing per atom in eV [defined in Eq. (1)] obtained from total energy calculations at a pressure of 10.5 Mbar and a temperature of 0 K. The symbols at specific values of x (number fraction of helium atoms) correspond to different ordered alloys in bcc and fcc crystal structures. The solid curve is a polynomial fit to both the bcc and fcc points.

diamond (or fcc) unit cell constructed from an fcc Bravais lattice with 4 atoms in the basis, all arranged along the cube diagonal; the net atomic packing for this structure is bcc. The label rhom refers to a bcc structure with a rhombohedral unit cell which contains a single atom of one type and its eight bcc nearest-neighbors of the opposite type. The curve in Fig. 1 is a polynominal fit to all of the calculated values (both bcc and fcc).

The calculated ΔH is a very slowly varying function of pressure. For all of the alloys considered, $\Delta H(x)$ is virtually unchanged between 5 and 10 Mbar. In addition, we find[12] that $\Delta H(x = 1/2)$ in the bcc-sc structure is nearly constant between 5 and 20 Mbar and that above 20 Mbar it decreases very slowly with increasing pressure (it has dropped by only 35% at 1000 Mbar). In our calculations pure helium metallizes at slightly less than 40 Mbar in the bcc structure (i.e., the energy gap between the 1s band and the 2p band goes to zero). However, we find that nothing striking happens to ΔH at 40 Mbar; it is still large (for $x = 1/2$ in the bcc-sc structure it has dropped by only 10% from the value at 5 Mbar) at this pressure and remains large at higher pressures.

The relatively large, positive values of ΔH in Fig. 1 imply that a high temperature must be reached before entropy dominates and the fully mixed phase becomes energetically favored over the coexistence of a helium-rich phase and a helium-poor phase. In order to directly address the question of phase separation, we construct a simple model which extends our TE calculations to nonzero temperatures. We consider only the ideal entropy of mixing. With this assumption we can calculate the Gibbs free energy of mixing,

$$\Delta G(x) = \Delta H(x) + k_b T\,[x \ln x + (1 - x) \ln(1 - x)], \qquad (2)$$

where k_B is the Boltzmann constant.

Through a series of common tangent constructions[12] we can determine the phase separation temperatures for all values of x. The results of this calculation, at a pressure of 10.5 Mbar, are shown as the solid curve in Fig. 2. This curve indicates that more than two phases can coexist in a small range of temperatures around 19,000 K.

We can estimate the additional uncertainty arising from our simplified treatment of thermal effects by a comparison with plasma models for which the equation-of-state of the solid and fluid are known accurately. We first calculate the phase separation temperatures, at a pressure of 10.5 Mbar, by applying the linear-mixing (LM) model[13] to a fit of the Helmholtz free energy obtained from the Monte Carlo simulations of Stringfellow et al.[14] for the one-component plasma (OCP). These results are plotted as the dotted curve in Fig. 2 (labeled OCP-LM). We next consider the ion-sphere model[15] which makes essentially the same approximations for the ion-ion

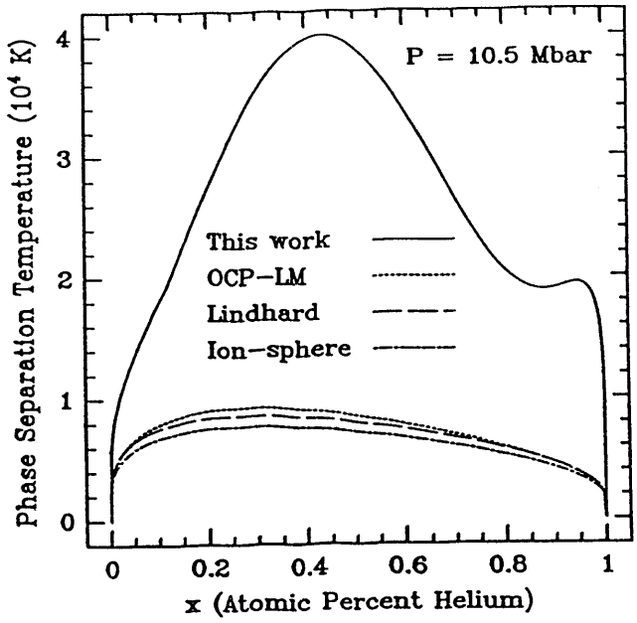

Fig 2. The solid curve is our calculation of the phase separation temperatures at a pressure of 10.5 Mbar [based on the results of total energy calculations and using Eq. (2)]. The critical (maximum) temperature is estimated to be T_c = 40,000 K ± 12,000 K at a critical concentration of x_c = 0.43. We also find the phase separation temperature at x = 0.07 (relevant to Jupiter and Saturn) to be T = 15,000 K ± 3000 K. The remaining curves are obtained from three different plasma models (see text).

interaction as in the OCP-LM calculation, but includes thermal effects exactly as we do in Eq. (2). The phase separation temperatures for this model, at 10.5 Mbar, are plotted as the dot-dashed line in Fig. 2 (labeled Ion-sphere). The difference between the ion-sphere and OCP-LM curves at x = 0.07 is very close to 1000 K and the corresponding difference in the critical temperature is approximately 2000 K. We take these quantities to be estimates of the uncertainty due to the simplified treatment of thermal effects in our calculation. Both plasma models may be compared to the Monte Carlo simulations of Hubbard and DeWitt[8] (at 10.5 Mbar) who used the Lindhard dielectric function to screen the bare Coulomb interaction between the ions (dashed curve in Fig. 2--labeled Lindhard). The Lindhard results are very close to both the OCP-LM and ion-sphere curves but very far from our calculations.

The primary conclusion obtained from the results of our TE calculations is that our phase separation temperature of 15,000 K± 3000 K for a 7% helium mixture confirms that the fluid interior of Saturn has phase separated, since the maximum temperature in the fluid is estimated to be only 10,000 K. Our phase separation temperature of 15,000 K is virtually independent of pressure in the range from 5 Mbar to 20 Mbar. The

estimated temperatures in the fluid interior of Jupiter range from 10,000 K near the surface to 20,000 K at the central core.[1] Thus our calculation predicts that phase separation has also begun in Jupiter. In view of this prediction, the fact that the current successful evolutionary models of Jupiter do not need to invoke phase separation may indicate a failure of these models. Alternatively, phase separation may have occurred too late in the evolution of Jupiter to provide a significant internal energy source up to the present time. In either case, new evolutionary calculations are needed to resolve this dilemma and to confirm that the inclusion of phase separation leads to a consistent model of Saturn.

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